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Biomass torrefaction for renewable energy: from physicochemical, bulk properties and flowability to future perspectives and applications

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ABSTRACT

18 Biomass and biomass wastes can be a source of renewable energy and fuels through valorization
19 in thermochemical processes. Torrefaction is a thermal pre-treatment often employed for
20 upgrading raw biomass. In addition to providing the status of current techniques used to
21 characterize raw and torrefied biomasses (in terms of their flowability and physicochemical,
22 thermal, and bulk properties), we comment on current applications with these bulk solids. The
23 limitations of current characterization methods are also discussed with a view to future scopes
24 with advanced techniques, particularly related to physicochemical properties. This review
25 underscores a lack of systematic studies focused on the importance of comprehensive
26 knowledge of raw and torrefied biomass properties to achieve better flowability, thereby
27 contributing to more efficient and cost-effective industrial processes. Indeed, among the 1320
28 literature papers evaluated in this review, 647 characterized the chemical/thermal properties of
29 raw and torrefied biomasses, while 254 considered physical/bulk properties, and only 11
30 assessed bulk solids' flowability. The scarcity of studies on flowability suggests that this
31 parameter has not been considered important by most researchers for the demonstration of
32 process feasibility. However, characterizing bulk solids flow behavior is critical for the proper
33 design of handling equipment and ensuring smooth plant operation, thus minimizing risks
34 associated with unforeseen expenses and prolonged time for process troubleshooting and
35 equipment retrofit. Moreover, even when the flowability was measured in the 11 papers, it was
36 difficult to compare results between studies as measurement techniques were not the same, nor
37 were the biomass type or torrefaction conditions. This highlights the need for future research
38 on the flowability of raw and torrefied biomass, intending to obtain more sound and broad
39 conclusions for the flow behavior of such heterogeneous materials, culminating in the
40 development of standardized protocols to improve biomass handling and processing at an
41 industrial scale. Besides, most of the studies available in the literature were based on small-
42 bench torrefaction units, producing only a few grams of torrefied biomass. This is generally a
43 limiting amount of material for complete assessment of flowability at different bulk conditions,
44 as well as to evaluate other important handling aspects at industrial scale, such as bulk solid
45 segregation, quality of fluidization, and so on. In future works, we also suggest evaluating other
46 heterogeneous feedstocks, such as municipal wastes or refuse-derived fuels, and performing a
47 complete characterization for the bulk solids to facilitate technical decision-making in
48 bioenergy and biofuels processes.

49

50 **Keywords:** Biochar, pyrolysis, powder handling, Hausner ratio, angle of repose, shear cell,
51 fluidization.

52 **1 Introduction**

53 The expansion of agricultural production and industrial processing can lead to an increase
54 in organic waste generation. In such a scenario, the valorization of biomass resources can
55 contribute to a circular economy by considering biomass's potential reuse or processing from
56 several feedstocks, including wood and forestry wastes, organic wastes, and agricultural
57 residues. The demand for sustainable energy solutions has directed significant research
58 attention towards biochar or torrefied biomass, a carbon-rich material produced from biomass
59 through processes like torrefaction.¹⁻⁴

60 Biomass residues are sustainable resources with large availability and easy access.
61 However, its use in raw form for thermochemical conversion processes is generally
62 compromised due to issues related to bulk solids handling and reduced energetic efficiency.⁵
63 This is a consequence of raw biomass properties, consisting of low bulk density, high moisture
64 content, reduced flowability, low calorific value, low energy yield, and high power
65 consumption for grinding. To make biomass profitable for large-scale operations, a
66 pretreatment step such as torrefaction is often necessary.

67 Figure 1 presents a simplified diagram describing dry biomass torrefaction. This partial
68 thermal conversion occurs for mild temperatures (generally within 200°C and 300°C), in the
69 absence of any oxidizing agents, and leads to the formation of three main products: volatile
70 matters either (i) condensable (steam and primary tars) or (ii) non-condensable (mainly CO and
71 CO₂) and (iii) a solid brown to dark residue called torrefied biomass. Note that other treatments
72 employing either “wet” or “steam” atmospheres can be also classified as torrefaction to obtain
73 an upgraded solid fuel, as described elsewhere^{6,7}.

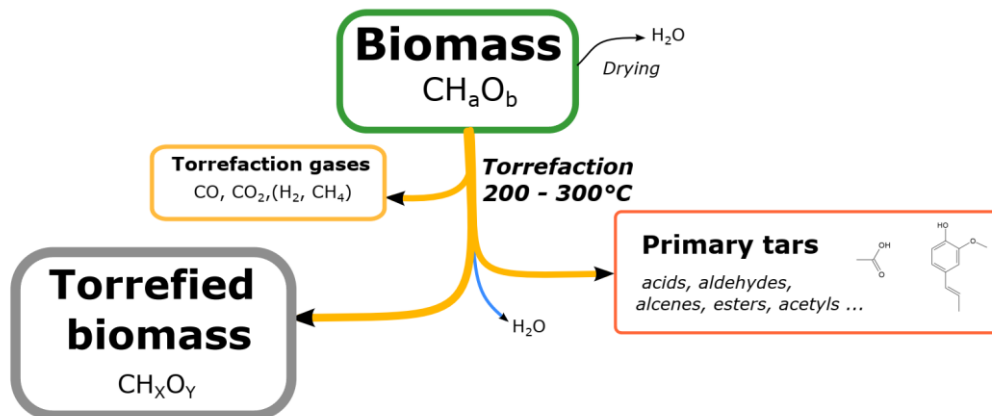


Figure 1. Diagram of dry biomass torrefaction.

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77 Industrially, two types of technology exist for dry biomass torrefaction according to the
78 heating method⁸. In the first type, the heat can be supplied to the reactor by direct contact
79 between hot inert gas and solids. This process mainly includes rotary drums, fluidized bed
80 reactors, and multiple hearth furnaces. In the second type, the heat can be applied indirectly to
81 the oven, by circulating, for instance, through a jacketed reactor. This process mainly includes
82 screw reactors and rotary drums.

83 The torrefaction severity (or degree of torrefaction) is often characterized by the solid
84 mass yield on a dry basis also called Anhydrous Weight Loss (AWL). For a given torrefaction
85 technology and particle size, it mainly depends on two operating conditions: the temperature
86 and the solid residence time. Indeed, a rise in these two parameters increases the degree of
87 torrefaction which leads to the formation of a browner and darker torrefied solid. For instance,
88 a typical mass yield for biomass torrefaction is 70% corresponding to an AWL of 30%.^{9,10}
89 According to the AWL value, it is then possible to i) significantly decrease the required energy
90 for grinding since the higher the AWL, the lower the energy required for grinding^{11,12}; ii)
91 enhance fluidization and flowability characteristics due to the improvement of particle size and
92 morphology, leading to more efficient handling operations;¹⁰ iii) produce a hydrophobic solid
93 with a low moisture content leading to long storage capacity¹³; iv) increase the biomass
94 durability, which decreases the biological conversion and leads to less decomposition risks;⁸ v)

95 increase the energy density with lower H/C and O/C ratios as the heating value is also increased
96 to approach that of coal^{2,14}.

97 Physical and chemical properties of torrefied biomass play a crucial role in determining
98 its suitability for many applications. For instance, the versatility of torrefied biomass extends
99 to energy applications, where it serves as a bridge between biomass and high-value products.
100 In energy conversion, the high carbon content and stability of torrefied biomass make it a
101 valuable precursor for producing activated carbon and other carbon-based materials. Its
102 potential in energy storage is highlighted by its application in supercapacitors and batteries,
103 where its porous structure and high conductivity are advantageous. Furthermore, the torrefied
104 biomass can be used as a fuel, either as a direct combustion fuel or as a feedstock for producing
105 syngas, biofuels, and bio-oil through gasification and pyrolysis.

106 Bulk and flowability characteristics of torrefied biomass are key for continuous operation
107 and process yield. According to Dai *et al.*,¹⁵ feeding waste biomass powders into
108 thermochemical reactors is commonly the most problematic operation of the entire reactor
109 system, because of blockages of pipes, feeding devices, and silo outlets. Issues arising from
110 poor powder flowability can occur under a range of flow and stress conditions. For example,
111 powder storage and discharge from silos often involves a quasi-static consolidated
112 configuration for the powders, and common flow issues consist of irregularity and interruption
113 of the flow of powders, rathole formation in funnel flow that can induce cross-contamination
114 and rotting of material accumulated in the silo, slip-stick flow that can endanger the silo, etc.
115 In contrast, processes such as fluidization and pneumatic transport involve a more dynamic and
116 loose packing condition¹⁶, for which typical flow issues consist of blockage of pipelines,
117 segregation and elutriation of finer particles, cross-contamination and rotting of accumulated
118 material, etc. Therefore, different flowability methods and indices have been developed over
119 the years to assess the flow behavior of a powder under different conditions of particle's

120 concentration, particle's relative movement, and their interaction with the system's boundaries
121 (*i.e.*, if free-surface or confined systems). A complete discussion in this sense is provided in
122 Section 3.3.1. Nevertheless, flowability data is key for the proper design of plant equipment
123 (*e.g.*, silos, solid feeders, mechanical/pneumatic transfer lines, etc.) and can be useful to ensure
124 smooth plant operation (*i.e.*, by monitoring flowability during operation as a quality parameter
125 to avoid flow instabilities and equipment blockage if bulk solid properties change), avoiding
126 unforeseen expenses and prolonged time for process troubleshooting and equipment retrofit.
127 However, these flowability properties are often mistakenly overlooked in research publications,
128 as if they were not necessary for demonstrating process feasibility.

129 Thus, this review paper aims to systematically evaluate the current state-of-the-art of
130 biomass torrefaction, with a focus on the methods used for the characterization of raw and
131 torrefied particulates, while proposing new fields of work based on our findings for advancing
132 sustainable energy solutions. Also to provide the significance of understanding the complete
133 properties of bulk materials, including their physical, chemical, thermal, bulk, flowability, and
134 fluidization characteristics. There are currently some reviews on the physical and chemical
135 characterizations of raw and torrefied biomass^{9,13,17,18}; however, none focused on the
136 characterization of flowability which is investigated in depth in our paper. The absence of
137 comprehensive reviews on flowability is a critical knowledge gap that must be addressed,
138 intending to guide future work that could lead to more efficient handling of biomass/biomass
139 waste, reduce waste, and improve industrial processing methods. In this paper, the discussion
140 is structured to cover several aspects of biomass torrefaction, as follows:

- 141 - The methodology used in this systematic review is presented in Section 2 and used in
142 Section 3.1 to obtain quantitative information on the number of papers published,
143 types of biomasses considered, and number of papers that investigated raw/torrefied

144 biomass characterization based on i) chemical & thermal properties, ii) physical &
145 bulk properties, and iii) flowability & fluidization properties.

146 - In Section 3.2, we describe briefly the current methods used for the physicochemical
147 characterization of raw and torrefied biomasses, while proposing other advanced
148 techniques for future work to fill some knowledge gaps.

149 - In Section 3.3, we present an in-depth analysis of the flowability and fluidization of
150 raw and torrefied biomasses, which is the core focus of our work. Flowability methods
151 are critically reviewed and key information about torrefaction conditions and
152 flowability data are extracted from the literature papers. The data is further grouped
153 for visualization and critically analyzed on which methods and biomass types are most
154 used, most productive countries, etc. Finally, we suggest future work to fill knowledge
155 gaps on flowability and fluidization linked to biomass torrefaction.

156 - In Section 3.4, we describe the main applications with torrefied biomass extracted
157 from the literature along with potential future trends.

158 - In Section 4, we propose final considerations on current and future literature, based
159 on the critical analysis provided in this article and our findings.

160

161 **2 Systematic review methodology**

162 The systematic review consisted of three stages and followed the PRISMA protocol
163 (Preferred Reporting Items for Systematic Reviews and Meta-Analyses),¹⁹ considering the
164 Scopus and Web of Science databases. As authors in the literature adopt different
165 nomenclatures for the biomass after the torrefaction process, the chosen search string was
166 “(‘biochar’ OR ‘biocoal’ OR ‘torrefied biomass’) AND ‘torrefaction’.” The research included
167 documents in English, and it was current as of March 20th, 2024. In the first stage, documents
168 from both databases were retrieved, and duplicates were removed. The gathered information

169 included title, publication year, authors, affiliation, and keywords. For each database, co-
170 occurrence clusters (network visualization) were prepared in VOSviewer® software (version
171 1.6.19) with the keywords of the selected articles.

172 In the second stage, there was a selection of papers within the scope of our research that
173 focused on biomass torrefaction, including some wastes that contain biomass, such as municipal
174 solid wastes, refuse-derived fuels, or solid recovered fuel. It was considered as out of scope
175 non-experimental research such as reviews and simulations as they do not provide new
176 information about bulk solid properties or characterization methods. For the selected papers,
177 we retrieved information regarding the type of biomass used for torrefaction to provide an
178 overview of the most studied feedstocks and their relevance in the context of a circular
179 economy. The remaining papers underwent individual qualitative analysis, categorizing them
180 as “yes”, “limited”, or “no” based on the availability of information about the physical,
181 chemical, and flowability characteristics of raw and torrefied biomass. Basic chemical
182 characterization should include proximate analysis, such as moisture, volatile matter, fixed
183 carbon, and ash contents, and ultimate analysis, involving the contents of carbon, hydrogen,
184 oxygen, nitrogen, sulfur, and other elemental compositions. In addition, the provision of
185 thermal characteristics should also be considered, such as mass and energy yields and heating
186 values. Physical and bulk characteristics involve particle size distribution, true and bulk density,
187 porosity, specific surface area, and morphological features, such as shape observed through
188 techniques such as scanning electron microscopy (SEM). Flowability characteristics include
189 static and dynamic angle of repose, Hausner ratio, Carr index, and flow function measured via
190 rheometers. Additionally, information on fluidization curves and permeability was also
191 considered.

192 In the third stage, we focused on studies that have conducted experimental approaches to
193 determine flowability characteristics. Additionally, we included studies from other reviews or

194 those cited within the selected papers that were relevant to our research scope but were not
195 retrieved in our systematic process. Flowability assessment of blends between torrefied biomass
196 and pulverized coal for co-processing operations was considered out of the scope of this review
197 paper, particularly if the flowability characterization was not performed for the raw biomass.
198 Some interesting papers on this subject can be found elsewhere.²⁰⁻²² For the final selection of
199 papers, we have provided information on the flowability or fluidization of torrefied biomass,
200 discussed their main characteristics and critically reviewed these studies. Finally, we explored
201 current applications of torrefied biomass and future perspectives, offering suggestions for future
202 research.

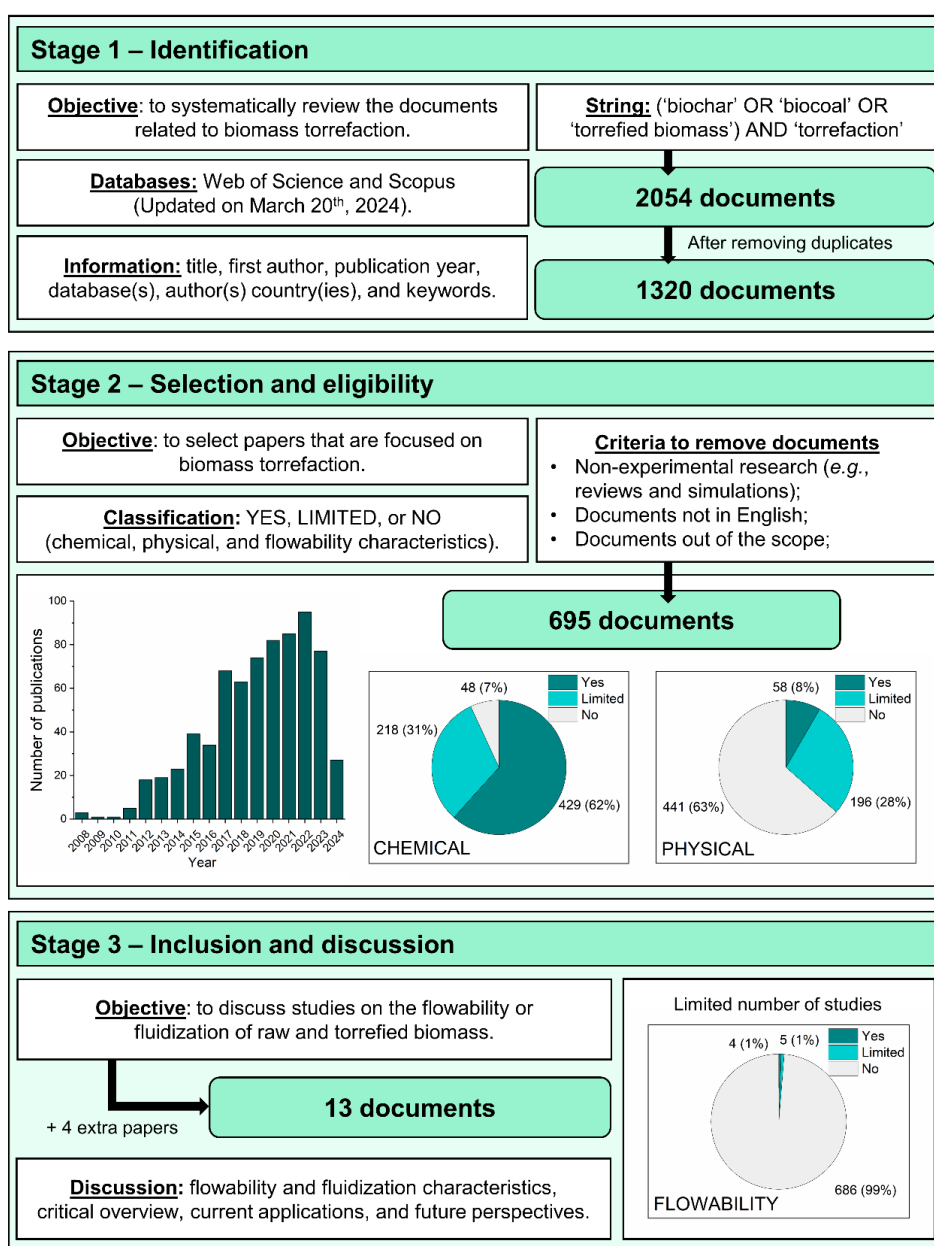
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204 **3 Results and Discussion**

205 3.1 Results of systematic review

206 Figure 2 presents a summary of the systematic review process and the selection of papers
207 for each stage. In the first stage, 2054 documents were retrieved, with 1143 from Web of
208 Science and 911 from Scopus. Figures S1 and S2 (Supplementary Information) present the co-
209 occurrence clusters with the keywords and affiliations of the selected articles from each
210 database. After removing duplicates, 1320 documents remained. In the second stage, 695
211 documents were selected based on our eligibility criteria. Considering the number of
212 publications per year, we observed an increasing trend, which aligns with the emerging interest
213 in this field of study. For instance, most papers (58%) were published in the last five years (74
214 in 2019, 82 in 2020, 85 in 2021, 95 in 2022, and 77 in 2023). Most papers presented complete
215 chemical characteristics of the torrefied biomass (62%) compared to those that presented
216 physical characterization (8%). More than half of the papers (63%) did not provide sufficient
217 physical and bulk characteristics of the torrefied biomass, which is a limitation for the

218 predictability and optimization of industrial operations with these materials. Additionally, the
 219 different characterizations provided in each research challenge the comparison across studies.
 220 A brief overview of the chemical and physical characterization is presented in Section 3.2.
 221 Finally, the systematic approach retrieved 9 papers that assessed the flowability or fluidization
 222 of raw and torrefied biomass. Additionally, 4 documents were found either within the selected
 223 paper's bibliography or in other reviews. Therefore, 13 studies were selected and critically
 224 discussed in Section 3.3.



225
 226 Figure 2. Systematic review summary: The search process and selection of the studies focused
 227 on the flowability of fluidization of raw and torrefied biomass.

228 From all papers selected in the second stage (n = 695), wood-derived biomass was
229 identified as the most common feedstock for torrefaction (n = 326). These papers reported the
230 repurposing of wood chips, pellets, sawdust, shavings, and stems. Some studies compared the
231 biomass torrefaction of softwood (*e.g.*, pinewood) and hardwood (*e.g.*, teakwood).²³
232 Additionally, other studies reused waste wood from furniture manufacturing as a recycling
233 alternative.²⁴ Several factors explain the predominance of studies on wood, including its
234 abundance and availability, high energy density, and homogeneous composition. Several
235 varieties of grasses and herbaceous plants were notable among the studies (n = 67), including
236 pruning and weeding residues, with *Miscanthus* spp. standing out (n = 11).

237 Agro-industrial residues constitute a significant group in the selection of feedstocks for
238 biomass torrefaction, contributing to a circular economy through waste minimization and the
239 repurposing of by-products from industrial processes.²⁵ Rice residues, including husks and
240 straws, are extensively studied (n = 65), followed by corn stalks, stover, straw, and corncob (n
241 = 55), and wheat straw (n = 30). These feedstocks are among the three largest cereal crops
242 worldwide,²⁶ ensuring abundant residues available for biomass torrefaction. Sugarcane bagasse,
243 leaves, and straw (n = 28) also play a crucial role in enhancing the energy balance of agro-
244 industries and supporting ethanol production. Additionally, residues such as shells and husks
245 from various nuts (*e.g.*, almonds, coconuts, walnuts, hazelnuts, and peanuts) are studied (n =
246 27). Other agricultural biomass includes oil palm trunk, kernel shells, mesocarp, and fronds (n
247 = 31); coffee residues such as spent grounds and husks (n = 23); cotton stalks (n = 14); olive
248 stones, cake, kernel, pomace, pips, and oil residue (n = 14); sunflower seeds, husks, and stalks
249 (n = 10). Barley (n = 6), camellia (n = 6), oats (n = 4), canola (n = 3), cassava (n = 3), pigeon
250 pea (n = 3), and soybean (n = 3) residues have also been studied.

251 Specific organic wastes such as fruit peels, seeds, shells, and stones, along with empty
252 fruit bunches have been studied for biomass torrefaction (n = 18). Municipal solid waste,

253 particularly its organic fraction, has been studied in 10 papers, and the torrefaction process can
254 produce a high energy density solid fuel.²⁷ Sewage sludge (n = 18), microalgae (n = 20) such
255 as *Chlorella vulgaris*, mushrooms (n = 7), refuse-derived fuels (n = 6), and textile waste (n =
256 4) are unconventional but efficient feedstocks for torrefaction. Poultry and chicken manure (n
257 = 15), as well as cattle manure (n = 7), are livestock residues that have been studied for
258 torrefaction. Additionally, 54 papers have explored other types of biomasses, including
259 polyamide biocomposites, distillation residues, tea residues, among others.

260

261 3.2 Chemical and physical characterization

262 During torrefaction, biomass undergoes several transformations, leading to some
263 chemical and physical modifications. First, the moisture is removed by the initial drying step
264 (up to 150°C), the second stage remains relatively stable within the temperature range of 137°C
265 –181°C, to remove chemisorbed water, followed by chemical transformations of the
266 hemicellulose, cellulose and lignin for temperatures range of 180°C up to 580°C, and releasing
267 volatile compounds and solids. The produced torrefied biomass is an upgraded solid that is an
268 alternative to fossil fuels²⁸ for energy storage and conversion systems.^{29,30} The torrefaction
269 process increases the calorific value of the biomass, resulting in a more efficient fuel. As the
270 torrefied biomass is easier to be ground into a fine powder, it could also be used as a pulverized
271 fuel. Torrefaction significantly enhances the physical and chemical properties of biomass,
272 transforming it into a more efficient and versatile fuel source.

273 Through detailed physical and chemical characterization, the quality and suitability of
274 biomass torrefaction for various applications can be assessed and optimized, promoting its
275 adoption in sustainable energy and industrial systems. While there is substantial literature on
276 biomass characterization, this section focuses on presenting the techniques used for such
277 characterization and provides a critical perspective on the existing gaps in information.

278 Although the chemical and thermal characterization are available in 647 papers
279 screened in this study, there are limited papers which present both chemical/thermal and
280 physical/bulk characterization. For this purpose, the systematic review procedure selected 58
281 papers that have analyzed the physical and bulk properties properly (Table S1), in which 36
282 papers (Table S2) include chemical and thermal characterization (see these tables in the
283 Supplementary Information). The objective of the next subsections is to describe the trends of
284 characterization methods used for torrefied biomass, in terms of chemical and thermal
285 properties (Section 3.2.1) as well as physical and bulk properties (Section 3.2.2).

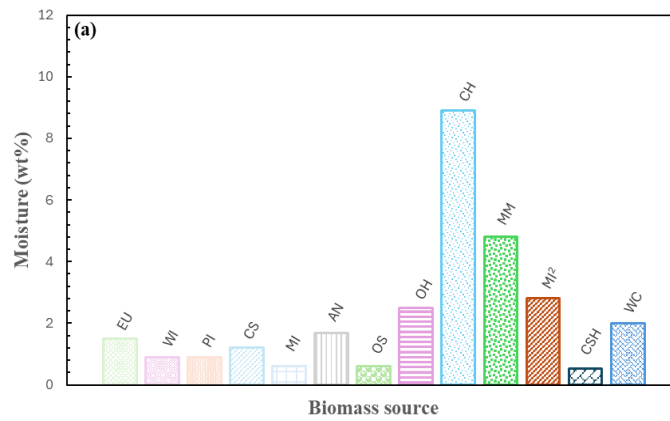
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287 3.2.1 Chemical and thermal characterization of biomass and torrefied biomass

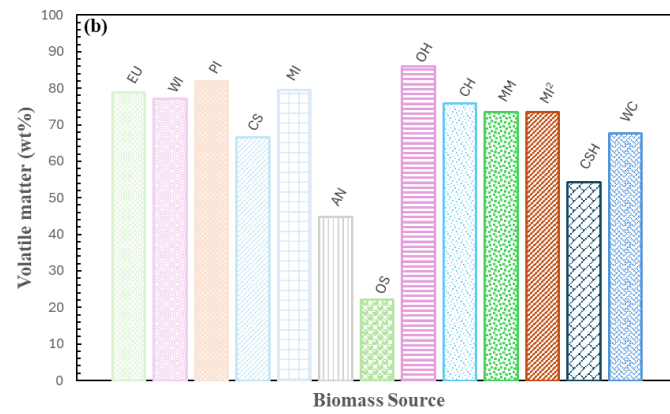
288 Chemical changes could be addressed by focusing on the changes in chemical
289 composition or/and structure. The proximate analysis involves determining the moisture,
290 volatile matter, fixed carbon, and ash content. The torrefied biomass may present a significant
291 discrepancy in the proximate analysis results, considering the different methods, such as those
292 from the American Society for Testing and Materials (ASTM) and thermogravimetric analysis
293 (TGA), and the process parameters (*e.g.*, heating rate, gas flow, temperature) used to obtain it.
294 The elemental composition related to carbon, hydrogen, nitrogen, sulfur, and oxygen
295 (CHNS/O) supports searching for elemental changes along the torrefaction process, such as the
296 amount of carbon which an increase is expected, enhancing the material's energy density^{6,7}.
297 The amount of CHNS/O may vary according to the source of biomass, and the ultimate analysis
298 is important for understanding biomass thermochemical conversion. Although the chemical
299 characterization plays a role, the intrinsic nature and recalcitrance³¹ of the biomass leads to a
300 considerable variation for ultimate and proximate analyses (see Table S2). The values vary from
301 biomass to biomass and even within the same type of biomass. Figure 3 addresses some

302 important information to illustrate the torrefied biomass composition and to highlight this
 303 statement.

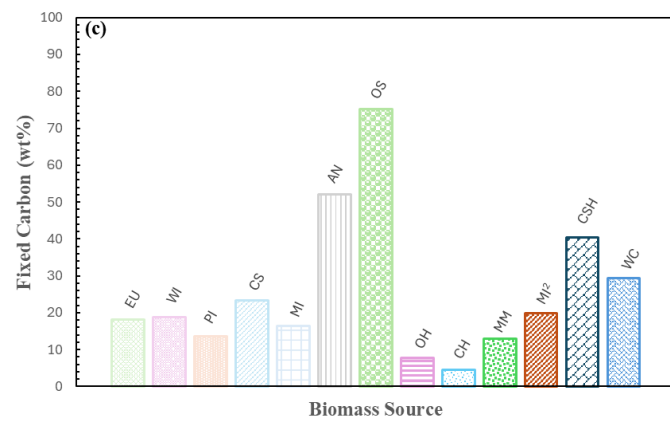
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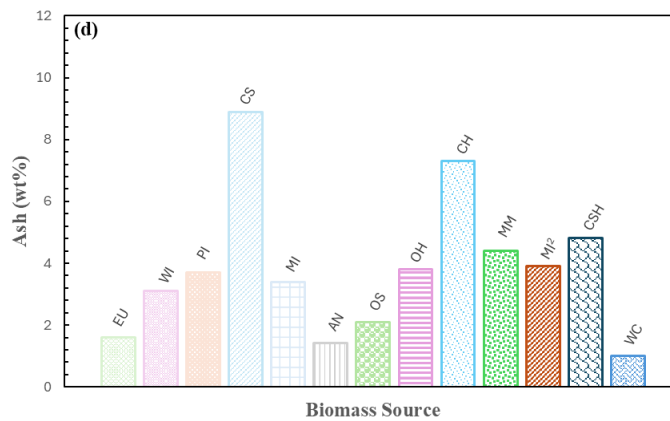
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307



308 Figure 3. Proximate analysis of different biomass sources. (EU: Eucalyptus, WI: Willow, PI:
309 Pine, CS: Corn Straw, MI: Miscanthus, AN: Acacia nilotica, OS: Olive stone, OH: Oat hull,
310 CH: Canola hull, MM: Mustard meal, MI2: Miscanthus, CSH: Camellia shell, WC:
311 Woodchips). (For reference see Table S2)³²⁻³⁷
312

313 The heating value of torrefied biomass is higher due to an increase in the carbon content
314 and due to the reduction of moisture. This could be evidenced by the TGA in which the stability
315 and decomposition are presented and defined by mass losses. TGA characterization can provide
316 insights into the combustion and pyrolysis kinetics, moisture, biomass degradation, endo and
317 exothermic reactions, ash content, which gives an overview of biomass behavior. The thermal
318 analysis will help to define future applications with torrefied biomass.

319 Another important characterization is Fourier Transform Infrared Spectroscopy analysis
320 (FTIR), which is used to identify changes in functional groups and chemical bonds, revealing
321 the extent of thermal degradation and the formation of new compounds. Some physicochemical
322 characterization, such as X-ray-based (*e.g.*, synchrotron-based scattering (SAXS), tomography,
323 Diffraction (XRD), Photoelectron Spectroscopy (XPS), fluorescence (XRF), Computed
324 Tomography (CT), etc.), need to be better defined in the context of torrefied biomass.³⁸⁻⁴⁰
325 Among the papers selected in this study (see Table S2), only five^{33,37,38,41,42} explored this type
326 of characterization to understand torrefied biomass. XRD provides a deep understanding of the
327 crystalline structure and can indicate the changes in the biomass's structural properties post-
328 torrefaction. XRD is important to characterize the type of carbon obtained during the process
329 (*e.g.*, graphite carbon atoms are arranged in layers, while graphene is only a single layer of
330 carbon atoms).⁴³ Combining the XPS technique with Raman Spectroscopy, it could be possible
331 to provide information regarding the type of carbon chemical bonds (*e.g.*, C=O, C-C, C-O) and
332 is possible to provide information about carbon defects, which could interfere with the energy
333 technologies applications. A full understanding of carbon sites could facilitate its application in
334 the context of energy technologies.^{44,45}

335 Nuclear Magnetic Resonance (NMR) has been used to provide detailed information about
336 the local chemical environment of carbon atoms, including the types of carbon-carbon bonds,
337 the presence of functional groups, and molecular structure⁴⁶. It can distinguish between
338 different types of carbon atoms, such as sp^2 , sp^3 , and hybridized carbons. However, this
339 technique has some disadvantages as it requires relatively large sample amounts, and the
340 analysis can be time-consuming. It is less effective for highly crystalline materials because it is
341 more focused on the local environment rather than long-range order.

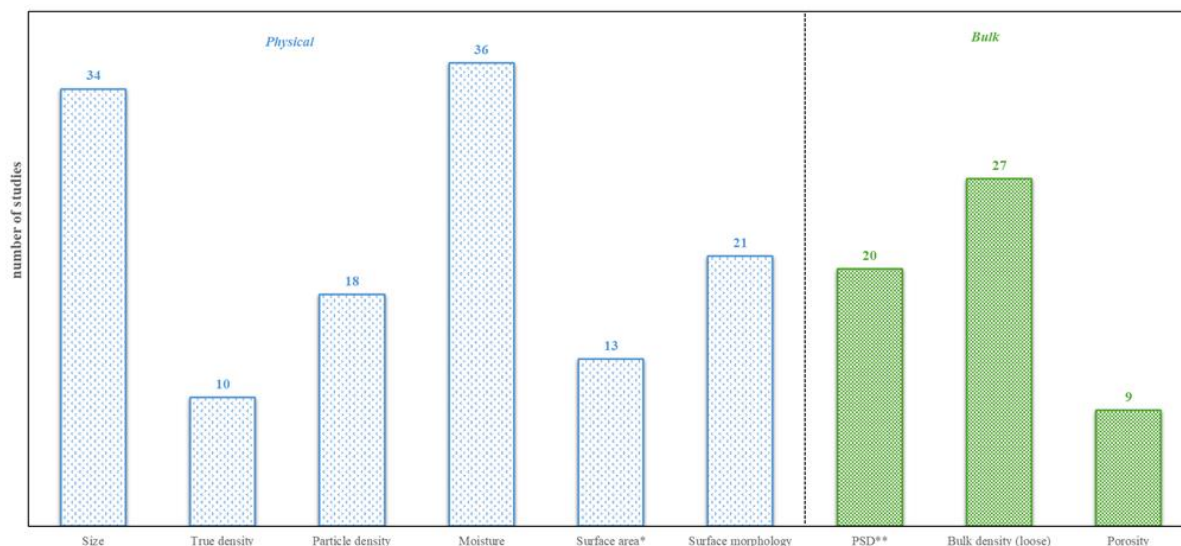
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343 3.2.2 Physical and bulk characterization of biomass and torrefied biomass

344 Knowledge about particle characteristics is important for the design of processes and
345 helps further optimizations. In terms of properties, it is needed to investigate those related to a
346 single particle, but also those that account for many particles and the voids within, known as
347 bulk properties. Overall, raw and torrefied biomass and biomass wastes present considerable
348 heterogeneity of particle and bulk properties, which is linked to different biomass types, nature
349 and structure but also to bulk solids processing methods and process conditions.

350 From the systematic review, 58 papers were selected with an appropriate characterization
351 of physical and bulk properties (Figure 4). In these papers, the most investigated biomass for
352 torrefaction is woody-derived ($n = 30$), followed by agro-industrial residues ($n = 23$),
353 wastewater ($n = 2$) and algae ($n = 1$), respectively. It is worth noting that some researchers
354 evaluated the impact of torrefaction from distinct sources of biomass. For this reason, the total
355 number of articles cited above exceeds the number of selected studies. Besides biomass
356 heterogeneity, the methods for biomass characterization are also a relevant factor that may lead
357 to some discrepancies in the measured properties values. For instance, 21 studies investigated
358 the particle size distribution (PSD) of raw and torrefied biomass, respectively. Among those,
359 18 studies assessed PSD by sieving out the particles and applying a gravimetric method while

360 fewer use laser diffraction ($n = 2$) and image analysis ($n = 1$). Different experimental procedures
361 may result in significant differences in particle size distribution, which makes comparison
362 between studies sometimes challenging.



363 Figure 4. Physical and bulk characterization of studies englobing biomass torrefaction.
364 (*Surface area is related to BET measurements, and it also involves mean pore size and pore
365 volume estimative; ** Particle size distribution. (For reference source see Table S1).
366
367

368 Bulk density is also a critical property since it mainly impacts process storage and
369 handling. Within the selected papers in this study ($n = 58$), 27 papers estimated values for bulk
370 density for raw and/or torrefied biomass. 22 papers estimated values for loose bulk density and
371 3 papers estimate values for tapped bulk density. Moreover, only 2 papers estimated values for
372 both loose and tapped bulk density. There is often a misunderstanding between the definition
373 and use of terms such as loose bulk density and tapped bulk density. The first one refers to the
374 not compressed state, while the second one is the density of the material after it has been
375 compacted.^{47,48} Considering the heterogeneous particle size, the shape of the biomass and the
376 process conditions (*e.g.*, pressure), clarifying this distinction is essential for handling it over
377 upstream and downstream processes. Loose and tapped bulk densities are also important for
378 estimating flowability parameters such as Hausner index and Carr's compressibility factor,
379 which are presented in the section 3.3.1.

380 Aligned with the discussion above, it is clear that bulk and physical properties should be
381 assessed carefully, and the methods adopted to estimate these properties must be in line with
382 the process phenomena. Bulk and particle properties affect flowability, which is an important
383 parameter for efficient handling and storage of bulk solids in different applications, particularly
384 for bioenergy and biofuels.

385

386 3.3 Flowability and fluidization characterization of biomass and torrefied biomass

387 From the systematic review procedure, a total of 13 papers were obtained to be critically
388 evaluated concerning the effect of torrefaction on biomass flowability characteristics ($n = 11$)
389 and fluidization characteristics ($n = 2$). The most used feedstock consisted of wood-derived
390 biomass ($n = 8$) followed by agro-industrial residues ($n = 4$), and one single occurrence for
391 waste textile fiber. The fluidization papers were published from 2012 to 2013, whereas those
392 addressing the flowability characteristics were published more recently, from 2017 to 2023.
393 With respect to flowability, the most productive countries were India and France with 4
394 occurrences each, followed by single occurrence papers from China, Mauritius, and Philippines.
395 Interestingly, among these papers the affiliation of the authors sometimes varied but it was
396 always within the same country. Scientific collaborations with experts from different
397 backgrounds in different countries should be promoted in the future. Finally, all the occurrences
398 for the fluidization papers were from Brazil ($n = 2$) with one co-author from France in one
399 paper.

400 In the following subsections, the flowability methods (Section 3.3.1) and selected papers
401 (Section 3.3.2) are critically reviewed, this latter in terms of main findings, torrefaction
402 conditions, type of bulk solid and flowability results interpretation. The flowability data from
403 all the papers are further grouped in Section 3.3.3 to discuss their trends and limitations. Finally,
404 a similar assessment is done for the selected fluidization papers in Section 3.3.4.

405 3.3.1 *Definition of flowability and critical overview of current methods*

406 By definition, a bulk solid is composed of solid particles and voids that are surrounded
407 by the environment or system boundary. The particles can have different physicochemical
408 characteristics (*e.g.*, various shapes, densities, sizes, chemical compositions, surface textures,
409 etc.), whereas the interstitial voids can be filled with different fluids of different densities and
410 viscosities. The environment or system boundary can have different arrangements, such as rigid
411 or moving walls of different textures, freeboards in which the bulk solid can be in contact with
412 fluids, or even regions of different pressures and temperatures. From this rationale, it is clear
413 that the environment impacts the bulk solids arrangement, and that its interaction combined
414 with the particularities of particles and voids characteristics, creates a complex system, which
415 is one of the reasons that developing a universal index to describe “flowability” is a challenging
416 task.

417 Bulk solid flowability can be defined as the likelihood of a particulate material to flow,
418 or in other words, as the potential for the particles to overcome the bulk inertia and have a
419 relative movement over other particles and its surroundings. There are different forces that
420 could prevent particles from moving, which can be of electrostatic origin, cohesive origin, or
421 even mechanical/interlocking⁴⁹. These have been well documented in the literature, although
422 direct measurement of these forces is still a challenge.^{50,51} Over the years, several indices were
423 proposed to assess, quantify, and discriminate the flowability of bulk solids, which have
424 different complexities but are fundamentally based on using a limited amount of material to
425 infer about the flow behavior of the bulk for a given process, generally at a much higher scale.
426 A critical review of current methods for flowability assessment is presented below.

427 Some classical methods involved the evaluation of flowability through simple powder
428 compaction tests, in which the degree of variation of the bulk density, between loose and
429 tapped/consolidated conditions, was used to indicate a better or worse flow behavior. In short,

430 powders that show a higher difference between these two conditions are classified with a worse
431 flowability, which is attributed to a higher intensity of electrostatic, cohesive, and/or
432 interlocking forces. The Hausner ratio (HR) is an index given by the ratio between
433 tapped/consolidated and loose conditions, whereas Carr's compressibility index (CCI) is given
434 by the standard difference between these variables.^{52,53} Powder compaction can be done by
435 vibration using automated or manual devices, for which the bed weight is the driven force for
436 bulk compaction, or even by using devices that impose consolidating pressures, in which there
437 is generally higher compaction of the powder bed. One important remark from these indices is
438 that they are measured under conditions of considerable high particle fraction, approaching
439 powders' packing limit; hence, the qualitative interpretation of the flowability might be
440 extrapolated, at best, to processes that undergo similar traits. Nonetheless, these indices are
441 mostly known for their quick and easy measurement that can be useful for a qualitative
442 differentiation between samples, and not necessarily for direct extrapolation to process
443 conditions. Some interesting works on these indices are presented elsewhere, for example
444 through modeling of the compaction procedure and via assessment of the impact of particle size
445 distribution, moisture content, ambient humidity and temperature, and so on.^{47,54-60}

446 Another classical index is the static angle of repose (α), which can be measured by the
447 fixed height or fixed diameter methods. The bulk solid is discharged by gravity through a funnel
448 to make a cone trunk on a flat or slightly concave surface. The literature rationale is that this
449 slope angle of the powder bed can be associated with higher or lower intensity of cohesion
450 forces. In short, a higher slope angle is an indication of worse flowability for the bulk solid.
451 Some authors claim that this index can be correlated with particle properties or even
452 dimensionless numbers, while others show that the angle of repose is only useful to plan storage
453 in a pile for bulk solids or predict the inventory in silos.^{61,62} It is worth noting, however, that
454 this test is performed under free-discharge of powders and that the pile is formed under loose

455 bulk conditions without any applied consolidation; hence, any extrapolation of flowability
456 information from this index should be done, at best, in such flowing conditions in the process.
457 One of the main advantages of this index is that it can be measured quickly and easily and can
458 give a first qualitative indication to discriminate the flowability of different samples,
459 particularly those samples that have considerable differences from each other. Some interesting
460 papers addressing the measurement of the angle of repose for bulk solids with different
461 properties can be found elsewhere.^{48,62-65}

462 The flow function coefficient (FFC) is another index that has been used for evaluating the
463 flowability of bulk solids, which is defined as the ratio of the major consolidation stress to the
464 unconfined yield strength. These parameters can be obtained in shear cells, which are devices
465 where the bulk solid is subsequently consolidated and sheared to derive the flow function at
466 each consolidation. As a consequence, the flowability indication can be evaluated at different
467 bulk solid conditions, corresponding to processes for which the bulk solids flow at
468 concentrations close to the packing limit and at different consolidation pressures. Note that this
469 is a different feature than the previous static angle of repose and compressibility indices, which
470 are measured for one condition generally at small bed weight consolidation. There are many
471 different commercial implementations of shear cells, from the precursor Jenike shear cell to the
472 Schulze ring shear tester, Brookfield powder flow tester, Freeman FT4 rheometer, and so on.
473 These implementations have different pros and cons concerning the consolidation range,
474 measurement speed, and degree of automatization. In general, the standard procedure of these
475 pieces of equipment leads to results with low variability under well-defined conditions (*e.g.*,
476 ASTM D6773-16).⁶⁶ Furthermore, the results of the shear cells allow not only for a qualitative
477 differentiation between the flow behavior of particulates but also provide valuable information
478 for the design of hoppers and silos since the cohesion, particle-particle, and particle-wall
479 friction parameters can be obtained from the tests. The effect of environment conditions, and

480 particle properties on the flow function has been extensively investigated in the literature,
481 including the comparison between the flowability indications given among the different
482 commercial shear cells.⁶⁷⁻⁷³ Some authors also commented on the limitations of shear cells to
483 measure flow properties of elongated or fibers type of biomass materials that are highly
484 compressible and severely entangled, which hinders the formation of a well-defined shear zone
485 and the attainment of steady-state flow condition.^{74,75}

486 More recently, rotating drums coupled with image analysis have been used to evaluate
487 bulk solids flowability at dynamic conditions. According to some authors, this device measures
488 dynamic flowability and can be understood as a shear cell at low consolidating stresses.⁷⁶ The
489 particulate is introduced in a drum that is continuously rotated while different characteristics of
490 the flow can be evaluated, mostly related to powder bed angle and the flow irregularity. It is
491 worth noting that depending on the drum rotating speed, different flow regimes can develop,
492 from unstable and intermittent to stable, cascading, cataracting, and avalanching modes, posing
493 more challenges for determination compared to the quasi-static indices (*i.e.*, α , HR, and FFC).
494 Furthermore, there is a greater interaction between fluid and bulk solids during the measurement
495 of this index, which can be challenging as the properties of solid and void fluid vary. For non-
496 cohesive materials, the angle that the particulate material makes with the horizontal under stable
497 flow is commonly named the dynamic angle of repose (β). However, for cohesive materials like
498 biomass, different parameters can be tracked between powder avalanches, such as the upper
499 and lower angle of stability, the frequency of avalanches, and so on.¹⁶ Generally, the higher the
500 dynamic angle of repose or angle of upper stability, the higher the interparticle frictional forces
501 in the powder, which is potentially an indication of worse flowability. The effect of bulk and
502 particle properties on the dynamic angle of repose has been evaluated in some works, however,
503 with considerably fewer data than shear cells.⁷⁷⁻⁷⁹ An indicative classification between powder

504 flowability as a function of dynamic angle or other dynamic quantities is still under
505 investigation.^{80,81}

506 From the rationale described in the previous paragraphs, the technique for flowability
507 assessment should be evaluated with caution and, more importantly, the test should be in line
508 with the actual process phenomenon under investigation, in which the bulk condition from the
509 process should be well reproduced, using a limited amount of material, during flowability tests
510 to allow for results interpretation and extrapolation at different scales.

511

512 *3.3.2 Description of selected papers for flowability*

513 In this section, the focus is to describe the main findings of the 11 papers that investigated
514 raw/torrefied biomass flowability and extract key information about torrefaction conditions,
515 type of bulk solid, flowability methods employed, and results interpretation by authors. A
516 chronological order is followed, when possible. The flowability data is summarized in Table 1,
517 along with the biomass type, torrefaction temperature and residence time, type of torrefaction
518 technology and unit scale, as well as other characterization methods used in each paper for
519 obtaining chemical, physical, bulk, and thermal properties.

520 **Table 1.** Flowability indices for the selected papers, consisting of Hausner Ratio (HR), Carr's Compressibility Index (CCI), static repose angle
 521 (α), dynamic repose angle (β), and flow function (FFC) at different pre-consolidation loads.

Authors	Biomass	T (°C)	Δt (h)	HR (-)	CCI (%)	α (°)	β (°)	FFC ^a (-)	FFC ^b (-)	FFC ^c (-)	FFC ^d (-)	Other information
Conag et al. (2017) ⁸²	sugarcane bagasse (SB)	0	0	1.49	33.00							- Fixed bed torrefaction at bench scale (9g of material). - Other tests performed: proximate analysis, calorific value, particle size, bulk density, combustion indices, and equilibrium moisture.
		250	0	1.44	30.33							
		250	0.25	1.42	29.33							
		250	0.5	1.32	24.00							
		250	0.75	1.32	24.00							
		250	1	1.25	19.67							
		250	1.25	1.33	24.67							
		300	0	1.44	30.33							
		300	0.25	1.32	24.00							
		300	0.5	1.31	23.67							
		300	0.75	1.37	27.00							
		300	1	1.39	28.00							
		300	1.25	1.35	25.67							
		350	0	1.35	25.67							
		350	0.25	1.27	21.33							
350	0.5	1.30	23.00									

		350	0.75	1.35	26.00	
		350	1	1.41	29.00	
		350	1.25	1.45	31.00	
Rago et al. (2018) ⁸³	waste textile fiber (WTF)	0	0	1.34	25.22	- Fixed bed torrefaction at bench-scale (65g).
		225	1	1.22	17.82	- Other tests performed: proximate and ultimate analyses, calorific value, FTIR, particle size, and bulk density.
		250	1	1.20	16.92	
		275	1	1.10	8.85	
		300	1	1.08	7.77	
		225	3	1.26	20.59	
		250	3	1.25	19.81	
		275	3	1.11	10.06	
		300	3	1.04	3.94	
Xu et al. (2019) ⁸⁴	fine soybean straw (SB)	0	0		60.9	- Fixed bed torrefaction at bench-scale.
		300	0.75		55.3	- Other tests performed: proximate analysis, SEM, particle size, PSD, bulk density, and shape factor.
	coarse soybean straw (SB)	0	0		61.2	
		300	0.75		55.3	
	fine corn straw (CS)	0	0		61.4	
		300	0.75		59.3	
	coarse corn straw (CS)	0	0		54.8	
		300	0.75		54.2	

	fine rice straw (RS)	0	0		64.9	
		300	0.75		56.8	
	coarse rice straw (RS)	0	0		70.0	
		300	0.75		68.2	
	fine rice husk (RH)	0	0		44.3	
		300	0.75		43.6	
	coarse rice husk (RH)	0	0		44.7	
		300	0.75		43.9	
Singh et al. (2019) ⁸⁵	pigeon pea stalk (PPS)	0	0	1.48	32.21	- Fixed bed torrefaction at bench-scale (6g).
		225	0.25	1.49	32.97	- Other tests performed: proximate and ultimate analyses, calorific value, FTIR, SEM, particle size, bulk density, combustion indices, and equilibrium moisture.
		225	0.5	1.46	31.55	
		225	0.75	1.44	30.65	
		250	0.25	1.39	27.93	
		250	0.5	1.38	27.77	
		250	0.75	1.38	27.66	
		275	0.25	1.38	27.75	
		275	0.5	1.37	27.31	
		275	0.75	1.36	26.51	
Singh et al. (2019) ³³	gum arabic wood (GAW)	0	0	1.29	22.48	- Fixed bed torrefaction at bench-scale (8g).
		252	1	1.21	17.35	- Other tests performed like previous work ⁸⁵

Singh et al. (2020) ⁸⁶	eucalyptus wood (EW)	0	0	1.44	30.43				- Fixed bed torrefaction at bench-scale (10g).	
		220	0.33	1.35	26.20				- Other tests performed: proximate and elemental analysis, calorific value, TGA, FTIR, XRD, BET, SEM, particle size, bulk density, particle density, combustion indices, and equilibrium moisture.	
		220	0.67	1.30	23.26					
		220	1	1.26	20.93					
		250	0.33	1.23	18.43					
		250	0.67	1.20	16.56					
		250	1	1.23	18.54					
		280	0.33	1.21	17.36					
		280	0.67	1.24	19.24					
		280	1	1.20	16.95					
Singh et al. (2020) ⁸⁷	gum arabic wood (GAW)	0	0	1.29	22.85	39.36			- Fixed bed torrefaction at bench-scale.	
		220	0.67	1.27	21.44	38.79			- Other tests performed like previous work ⁸⁶ except for XRD, BET, but including contact angle, and ICP-MS.	
		250	0.67	1.21	17.35	36.15				
		280	0.67	1.18	15.86	32.51				
Pachon-Morales et al. (2019) ⁸⁸	poplar wood (PW)	0	0				5.2	4.6	3.9	- Fixed bed torrefaction at bench-scale (15g).
		220	1				5	4.4	4.2	- Other tests performed: PSD, shape factor, and bulk density.
		220	5				4.8	4.9	4.1	
		250	1				5.3	5	5.1	
		250	5				6.9	6	6.3	
		280	1				6.6	6.3	5.7	

		280	5		9.2	9.4	8.5		
	spruce wood (SW)	0	0		3.5	3.9	3		
		220	1		3.7	3.8	3.4		
		220	5		4.9	4.5	4.1		
		250	1		4.7	4.8	4.2		
		250	5		6.5	7.4	6.2		
		280	1		6	6.2	6.3		
		280	5		5.8	8.4	11		
Pachon-Morales et al. (2020) ¹⁶	coarse poplar wood (PW)	0	0	146.3	7.0	10.0	8.6	- Fixed bed torrefaction at bench-scale (450g).	
		240	1	148.2	9.3	7.3	5.4	- Other tests performed like the previous work ⁸⁸	
		280	1	153.3	10.4	6.0	4.9		
	fine poplar wood (PW)	0	0	149.1	5.6	5.8	6.0		
		240	1	146.2	4.6	4.3	4.1		
		280	1	151.1	5.8	4.6	4.1		
Thevenon et al. (2021) ⁸⁹	mix of spruce/pine wood (SPW)	0	0	60	5	14.7	5.9	10	- Six-hearth furnace for torrefaction at pilot-scale (420kg).
		250	0.92	61	9.6	16.4	18.7	17.3	- Other tests performed: elemental analysis, calorific value, PSD, and shape factor.
Rajaonarivony et al. (2023) ⁹⁰	wheat straw (WS)	0	0	36.10					- Continuous screw reactor for torrefaction at pilot scale (2.92 kg/h). - Other tests performed: biochemical composition, AFM, TGA, NMR, calorific value, PSD, and shape factor.
		280	0.17	32.10					

522 Note: FFC^a was measured at pre-consolidation loads of 2-2.7 kPa, FFC^b at 5-5.3 kPa, FFC^c at 7.9 kPa, and FFC^d at 10-10.5 kPa.

523 Conag *et al.*⁸² investigated the torrefaction of sugar cane bagasse of 0.56 mm using about
524 9 g of material in a fixed bed reactor at different temperatures (*i.e.*, 250°C, 300°C, and 350°C)
525 and residence times (*i.e.*, 15, 30, 45, 60, and 75 min) under minimized oxidative atmosphere.
526 The authors evaluated the flowability via HR and CCI, indicating that the torrefied biomass had
527 relatively lower indices than the raw biomass, meaning better flowability but also a worse
528 compaction for these samples. The qualitative indication for the raw sample was “very poor”
529 and “passable” for the torrefied powder. Another finding was that a torrefaction time beyond
530 60 min did not further improve the fuel characteristics of torrefied biomass, in terms of energy
531 density and high heating value.

532 Rago *et al.*⁸³ performed the torrefaction of waste textile fibers of about 9 mm mesh size
533 at different temperatures (*i.e.*, 225°C, 250°C, 275°C, and 300°C) for 1 and 3 h in a batch reactor
534 with about 65 g of material. The flowability was assessed via the Hausner ratio and
535 compressibility index, generally showing a better flowability as the torrefaction severity (*i.e.*,
536 higher temperature and/or higher treatment time) was increased. The initial fibers were highly
537 fibrous and fluffy, corresponding to the highest bulk density compared to the torrefied material,
538 to which the characteristics of a more powdery, less fluffy, and rather porous material were
539 attributed.

540 Xu *et al.*⁸⁴ evaluated the flowability of four milled biomass powders (soybean straw, corn
541 straw, rice straw, rice husk) before and after torrefaction in a lab-scale fixed bed reactor
542 operated at 300°C for 45 min (amount of produced powder not indicated in the paper). The
543 flowability was assessed via static angle of repose (fixed bed method) for two sieve size ranges
544 of 250-380 µm and 380-550 µm. Overall, better flowability was observed for some torrefied
545 samples (soybean and rice straws), particularly for the finer particles, while for others (rice
546 husk, corn straw) the angle of repose remained almost identical even with the torrefaction
547 pretreatment. Moreover, although torrefaction can help in improving the flowability, it cannot

548 change the relative relationship between the flowability of different biomass powders. Finally,
549 through compressibility tests on raw milled biomass powders, the authors stated that the
550 Hausner ratio/compressibility index cannot always characterize their flowability compared to
551 the angle of repose, as a consequence of biomass nature (*i.e.*, specific internal structures,
552 flexibility, and shape). Note that the compressibility tests were not evaluated for the torrefied
553 sample.

554 Singh *et al.*⁸⁵ investigated the dry torrefaction of pigeon pea stalks of sieve sizes between
555 0.7 and 1.2 mm under different temperatures (*i.e.*, 225°C, 250°C, and 275°C) and residence
556 times (*i.e.*, 15, 30, and 45 min) using samples of about 6 g in a batch furnace. The authors
557 verified a decrease in the solid yield with increasing torrefaction severity, which was defined
558 by higher residence times and temperatures in the reactor. The flowability assessed via HR and
559 CCI was improved by increasing the torrefaction severity, being classified as “very poor” for
560 the raw sample and “poor” for the condition of 275°C and 45 min. Nevertheless, the authors
561 state that a low temperature of 225°C at any of tested residence times had no effects for
562 flowability improvement. Finally, the torrefied biomass showed a higher internal porosity and
563 lower bulk density (about 18% lower than raw biomass), tended to absorb less water, and had
564 reduced structural strength with more surface cracks that in turn helps in reducing crushing and
565 compaction power consumption.

566 In a following paper, Singh *et al.*⁸⁶ evaluated the torrefaction of 8g eucalyptus wood of
567 sieve sizes between 0.7 and 1.2 mm using the same reactor setup at similar temperatures (*i.e.*,
568 220°C, 250°C, and 280°C) and residence times (*i.e.*, 20, 40, and 60 min). Similar conclusions
569 were drawn from the experiments, consisting of an improvement of the flowability via HR and
570 CCI as the torrefaction severity was increased. At the limits, the raw sample presented a “poor”
571 flowability that was transformed into “fair” for the sample treated at 280°C and 60 min.

572 Singh *et al.*³³ experimentally evaluated the dry torrefaction in a fixed bed reactor, by using
573 about 10g of gum Arabic wood powders in each test, with a sieve size from 0.7 to 1.2 mm. They
574 evaluated various conditions for torrefaction temperature (from 220°C to 280°C), residence
575 time (from 20 to 60 min) and heating rate (from 5°C to 15°C/min), aiming at obtaining optimum
576 conditions for maximization of torrefied biomass high heat value and energy yield through
577 statistical Response Surface Methodology (RSM). The flowability was evaluated via HR and
578 CCI for the raw and torrefied sample of highest yield (*i.e.*, 252°C, 60 min, and 5°C/min),
579 showing an improvement of flowability from “fair” to “good”, respectively.

580 In a following paper, Singh *et al.*⁸⁷ evaluated the torrefaction of gum Arabic wood at
581 similar residence times (*i.e.*, 20, 40, and 60 min) and temperatures (*i.e.*, 220°C, 250°C, and
582 280°C), obtaining the optimum tradeoff between higher heating value and energy yield for
583 250°C and 40 min. The flowability was evaluated via static angle of repose, HR and CCI for
584 the raw sample and three other torrefied ones at 40 min and all temperatures. There was a
585 general improvement in the flowability indices, from the raw sample being classified as
586 “passable” and the torrefied at the highest temperature as “good”. It is worth mentioning that
587 the qualitative classification among different indices did not match perfectly, which is a
588 common finding in the literature and agrees with the rationale from Section 3.2.1, illustrating
589 that different flowability indices describe the flow behavior at different bulk flow conditions.
590 Also noteworthy is the use of advanced physicochemical characterization techniques in these
591 last two studies, such as FTIR, XRD, BET, and ICP-MS.

592 Pachón-Morales *et al.*⁸⁸ investigated the torrefaction of poplar and spruce wood chips of
593 1.5 mm of particle diameter at three temperatures (*i.e.*, 220°C, 250°C, and 280°C) and two
594 residence times (*i.e.*, 1 and 5 h) in a batch furnace containing about 15 g of material. Under
595 similar torrefaction intensities, Spruce presented higher formation of fines (particles below 100
596 µm) compared to poplar, which was attributed to distinct anatomical structures for the two

597 biomasses that lead to different fracture pathways. Besides, a gradual shifting of size and shape
598 distributions towards finer and rounder particles was verified when increasing the torrefaction
599 intensity. The flowability for all powders was evaluated with the Schulze ring shear tester at
600 consolidations of 2, 5, and 10 kPa. A significant improvement of the flowability was verified
601 as the torrefaction intensity was increased, from “cohesive” for the raw samples to nearly “free
602 flowing” for the condition of 280°C during 5 h. Moreover, the authors provided a
603 comprehensive discussion to state that the improvement of flowability was mostly related to
604 the rounder shape and sharper surface of torrefied biomass, resulting from a loss of resilience
605 of the fibrous structure of raw wood due to torrefaction, and not by the changes in the particle
606 size or size span.

607 In a following study, Pachón-Morales *et al.*¹⁶ focused on the torrefaction of poplar wood
608 chips at 240°C and 280°C for 1 h in a batch furnace using samples of about 450 g. After grinding
609 and sieving, the authors evaluated the flowability of raw and torrefied powders of two different
610 particle-size distributions through techniques of shear cell and rotating drum, representatives of
611 compacted and free-surface stress states, respectively. Overall, intense torrefaction improved
612 the flow behavior of powders, both under free-surface and consolidated conditions. Fine, mildly
613 torrefied powders showed the worst flow behavior, whereas the intensively torrefied coarse
614 sample has the best flowability. Note that for some conditions the flowability of raw biomasses
615 was better than the corresponding torrefied sample, indicating potentially that this pretreatment
616 does not always improve the flow behavior. This highlights the need for systematically
617 measuring the flow properties to draw best conclusions for ease handling and storage
618 operations.

619 Thevenon *et al.*⁸⁹ investigated the effect of torrefaction at pilot scale for wood chips,
620 consisting of a mix of 70% spruce and 30% pine, at the temperature of 250°C and residence
621 time of 55 min. The raw sample contained a moisture content of 52% wet basis, which was

622 initially dried in a pilot scale belt dryer to about 18% wet basis, and subsequently torrefied in a
623 continuous vertical six-hearth furnace (10 m in height and 1.82 m in diameter) to produce 420
624 kg of torrefied wood. Both raw and torrefied wood chips were ground with a knife mill
625 generating powders samples with mean sieve size of 662 and 264 μm , respectively. Torrefaction
626 not only reduced the grinding energy costs but also decreased by half the particle diameter,
627 which is related with the weakening of the particle structure due to torrefaction. The flowability
628 was evaluated with a rotating drum and Schulze shear cell representing bulk conditions of loose-
629 dynamic and consolidated-static states, respectively. With respect to the dynamic avalanche
630 angle, the raw and torrefied samples presented similar mean values of 60° and 61° , respectively,
631 however, the first had a pronounced erratic flow behavior evidenced by standard deviation of
632 38° , while the flow for the torrefied sample was more homogeneous with a deviation of 8° .
633 Concerning the shear tests, the raw sample was classified as a “easy-flowing” material, while
634 the torrefied was “free-flowing”, with the flowability change majorly attributed to the
635 smoothing of particle surfaces as the aspect ratio of the particles remained almost stable in their
636 work. Overall, the authors concluded that there was an improvement in the flowability, under
637 both loose and consolidated states, due to the torrefaction treatment.

638 Rajaonarivony *et al.*⁹⁰ performed the torrefaction of wheat straw in a pilot scale screw
639 reactor at 220°C and 280°C , feed rate of 2.92 kg/h, and residence time of 10 min. Torrefied and
640 non-torrefied biomass were ground in a stirred ball mill to obtain fine powders in the range of
641 3 to 120 μm . Torrefaction at the low temperature of 220°C did not result in significant changes
642 neither in biomass physicochemical and structural properties nor in their grindability.
643 Nevertheless, at 280°C there was 50% better grindability compared with the raw sample, but
644 also with higher tendency for agglomeration of fines, forming hard agglomerates in the range
645 of 200-600 μm . The agglomeration phenomena originated by torrefaction was attributed to
646 modifications of particle surface properties that make certain chemical groups more accessible

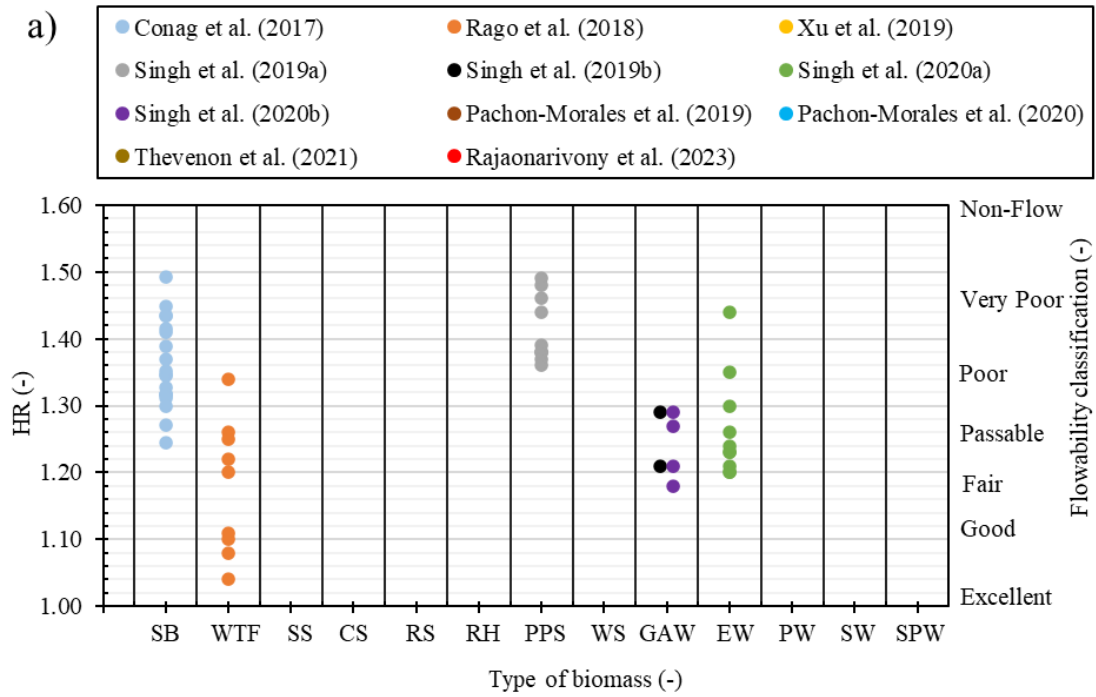
647 to form stronger interactions with others, but it requires further investigation. The flowability
648 was evaluated through compressibility index with applied pressure of 15 kPa and with FT4
649 rheometer for cohesion measurement. Overall, the flowability was improved from “very poor”
650 for the raw sample to “poor” for the torrefied powder at 280°C, while the cohesion was also
651 reduced by half in the latter. Torrefaction process makes straw particles more brittle, which
652 decreases their deformability and compressibility of the powder. Moreover, the authors state
653 that even a small improvement of the compressibility index (measured at quasi-static
654 conditions) would represent huge gains on powder handling as they can be associated with
655 significantly less mechanical forces to initiate the movement of particles. Besides, the cohesion
656 reduction in the torrefied powder was interestingly associated with the degradation of
657 arabinoxylans, a non-cellulosic component responsible for interparticle interactions of cohesive
658 nature.

659

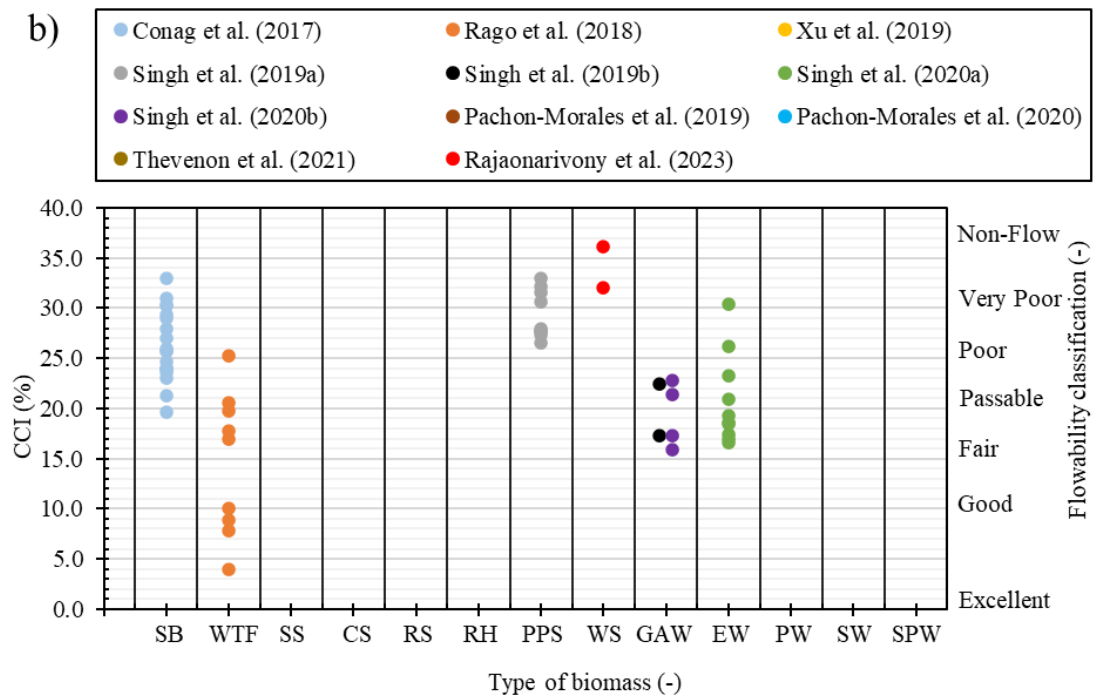
660 *3.3.3 Grouped flowability data and results discussion*

661 In this section, the flowability data gathered in Table 1 are discussed in more detail to
662 reveal the current status of the literature and visualize the gaps that need future studies. The
663 flowability indices considered were the HR, CCI, α , β , and FFC at different pre-consolidation
664 loads, for which an overview was done in Section 3.3.1. Dispersion plots were produced for
665 each flowability index as a function of the investigated biomasses, a total of 13 feedstocks. In
666 Figure 5, the data points are colored based on the 11 literature references for flowability, while
667 no occurrence of data points for a given biomass means that it has not been characterized with
668 the corresponding flowability technique. At the right axis, the commonly used flowability
669 classifications from the literature were applied for HR⁵², CCI⁵³, α ⁷⁶, and FFC⁶⁷. Note that a
670 classification for β is not yet available in the literature, as detailed in Section 3.3.1.

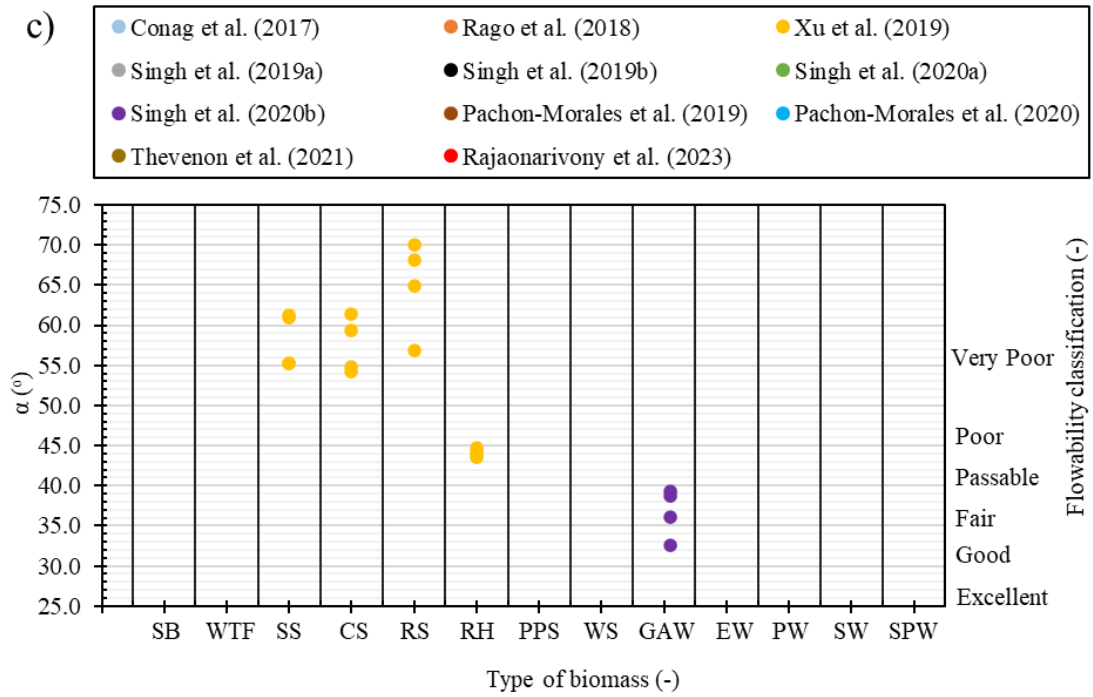
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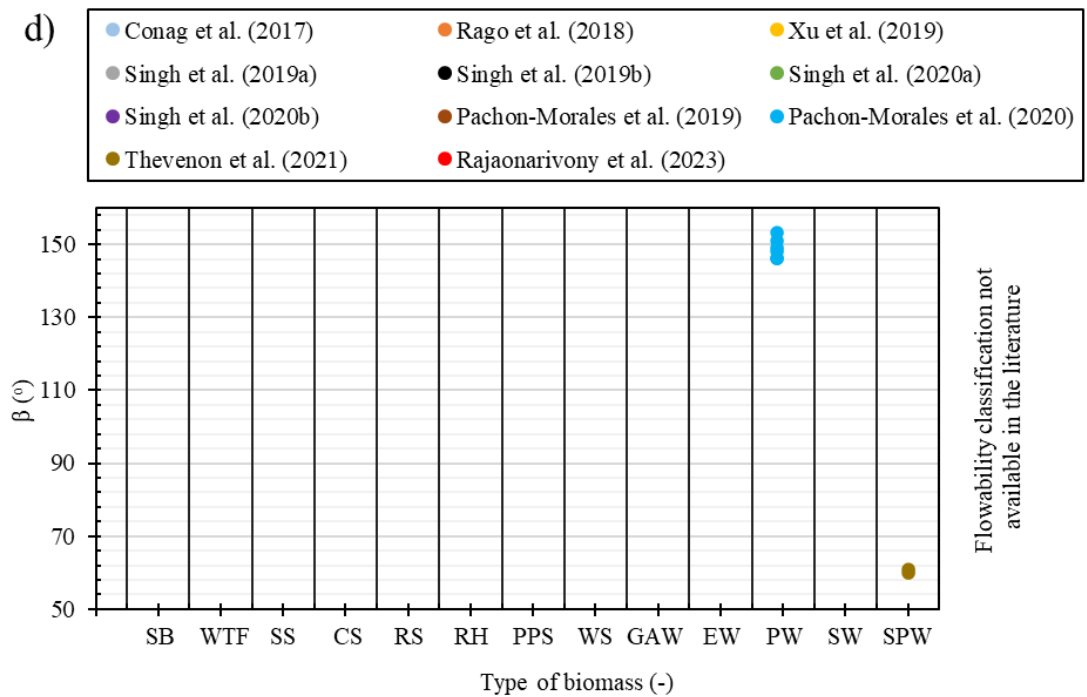
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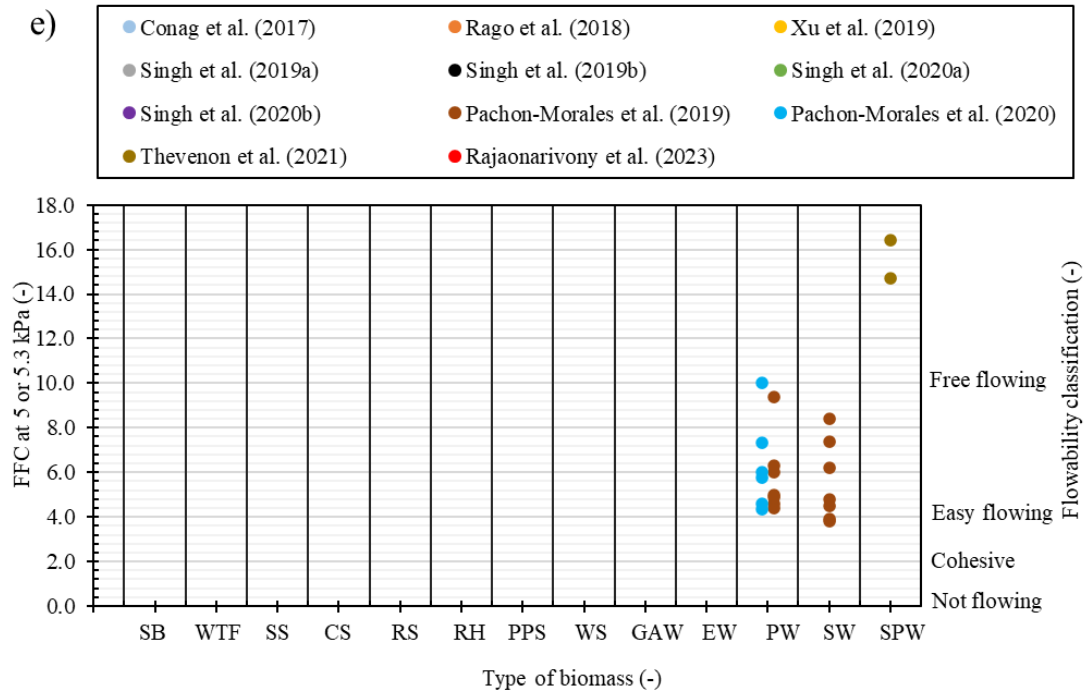
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675



676 Figure 5. Flowability indices as a function of different investigated biomass in the literature:
 677 a) HR, b) CCI, c) α d) β , and e) FFC. The points are colored based on the literature reference.
 678
 679 No occurrence of data points for a given biomass means that it has not been characterized
 680 with the corresponding technique.
 681

682 Overall, the classical indices derived from powder bed compaction, HR, and CCI, were
 683 those mostly used to assess raw and torrefied biomass flowability. Data dispersion in Figures
 684 5a and 5b revealed similar trends for both indices including similar flowability classification,
 685 as expected, since both are based on the same experimental unit and raw data. In total, 5
 686 feedstocks were characterized via HR with only one (*i.e.*, gum arabic wood) being evaluated
 687 by two different studies. Similarly, for the CCI, 6 feedstocks were assessed with only gum
 688 arabic wood being evaluated by two papers. Depending on the torrefaction treatment severity
 689 and feedstock, HR ranged from 1.10 (excellent flowability) to 1.49 (very poor). Similarly, CCI
 690 ranged from 3.9% (excellent) to 36.1% (very poor).

691 In Figure 5c, the static angle of repose was measured for 5 feedstocks in two literature
 692 papers, highlighting a considerable variation of flowability classification, from 32.5° (good
 693 flowability) to 70° (very poor), depending on the biomass sample and pre-treatment intensity.

694 It is worth noting that both papers used different techniques to measure the angle of repose,
695 consisting of fixed diameter and fixed height, which makes a direct comparison difficult and
696 establishes a warning for future studies. A similar comment can be attributed to Figure 5d, in
697 which only two papers considered the rotating drum technique, and yet the criteria used for
698 measuring the dynamic angle of repose was different among them, which explains the scattered
699 data and prevents comparison of results.

700 In Figure 5e, shear cells were used to assess the flowability of raw and torrefied wood
701 powders in three papers, for which the FFC (at a pre-consolidation load around 5kPa) ranged
702 from 3.8 (easy-flowing) to 16.4 (free-flowing). The data trends for FFC under different
703 consolidation loads can be observed in Table S3 (Supplementary Information). One of the
704 advantages to use of shear cells for flowability classification is that the parameters for silo
705 design, such as angles of internal friction and particle wall, can be made available from the
706 same set of experiments. Coincidentally, all papers used the Schulze ring shear tester for the
707 flowability data from Figure 5e; hence, it would be interesting in the future to compare these
708 results with other commercial implementations of shear cells available in the literature. An
709 illustration for this comparison was done previously for raw biomass.⁴⁹

710 Based on the previous flowability dataset (Table 1 and Fig. 5) and the overview of current
711 works associated with biomass torrefaction (Section 3.3.2), we found the following gaps and
712 potential fields for future work:

- 713 - None of the studies on biomass torrefaction provided a complete picture of flowability
714 using all available techniques, corresponding to different bulk flow conditions. In
715 addition, coupling this with a full characterization of physicochemical, thermal, and
716 bulk properties and techno-economic feasibility studies is pressing to improve the
717 performance of thermochemical processes and the circular economy of biomass and

718 biomass residues. Noteworthy is the use of advanced characterization techniques that
719 were employed in only 3 out of 11 works (see Table 1 for details).

720 - The comparison of results between the 11 studies was challenging as the flowability
721 measurement techniques were not always the same, nor were the torrefaction
722 conditions or the biomass type (which also implies quite different physicochemical
723 and bulk properties: size, shape, surface roughness, bulk density, etc.). Consequently,
724 current literature results are difficult to be applied elsewhere. Further works are
725 needed, under similar flowability testing conditions and under a determined range of
726 biomass properties, in order to obtain more sound and broad conclusions that allow
727 for the establishment of standardized protocols for handling heterogeneous
728 feedstocks, such as raw/torrefied biomass, at different process scales.

729 - None of the studies investigated the effect of electrostatic forces or evaluated in-depth
730 interlocking effects or cohesion induced by moisture absorption, which prevents
731 further discussions in this regard. The current focus of work was to report an
732 improvement or not in the flowability, for very specific combinations of a given
733 biomass and torrefaction condition. Further works centered on raw/torrefied biomass
734 flowability assessment under higher intensity of electrostatic, cohesive & interlocking
735 forces would be useful, not only for the bulk solids handling research field but also
736 for more assertive applications with heterogeneous feedstocks.

737 - Most of the studies in the literature (9 out of 11) were based on small-bench
738 torrefaction units, producing only a few grams of torrefied biomass. This is generally
739 a limiting amount of material for complete assessment of flowability at different bulk
740 conditions, as well as to evaluate other important handling aspects at industrial scale,
741 such as bulk solid segregation, quality of fluidization, and so on. Thus, we suggest

742 this as a scope for future work aiming at improving the success of biomass torrefaction
743 at different unit scales.

744

745 3.3.4 Description of selected papers for fluidization

746 Rousset *et al.*⁹¹ investigated the fluidization characteristics of wood chips of three size
747 distributions, corresponding to raw and torrefied biomass at 200°C and 15 min. Overall, a
748 higher pressure drop was observed for the raw biomass samples at the same gas velocity, which
749 was attributed to the weakening of the torrefied sample. In a subsequent work, Rousset *et al.*⁹²
750 evaluated the grindability by fluidization of three wood chips samples with different size
751 distributions, before and after torrefaction for 15 min at 210°C, 240°C, and 270°C. The
752 fluidization tests were performed with 450 g of material and for 20 min at an increasing gas
753 velocity up to 3 m/s. With respect to the raw samples, only big particles higher than 1.6 mm
754 showed a reduction in size by fluidization. On the other hand, torrefaction had a significant
755 impact on particle grindability by fluidization for all sizes from 0.28 to 1.6 mm. The size
756 reduction was also correlated with the torrefaction severity, with higher treatment temperatures
757 originating finer particles. Nevertheless, further studies are needed in this field to show the pros
758 and cons of biomass torrefaction for fluidized bed technologies, particularly by studying more
759 complicated biomass residues (*e.g.*, refuse-derived fuel or municipal waste) and by
760 investigating other important phenomena in fluidization, such as gas-solid contacting, particle
761 segregation, particle attrition, and particle entrainment.

762

763 3.4 Current applications of torrefied biomass and future perspectives

764 Not all retrieved studies on the flowability and fluidization of torrefied biomass
765 highlighted the potential applications of their findings. Most of these studies primarily focused

766 on enhancing the energy potential of biomass. Proper physical and bulk characterization is
767 essential to improve the handling and stability of torrefied biomass, an important step to scale
768 up the experiments to field applications. Additionally, there are environmental benefits of
769 torrefied biomass, including soil amendments and remediation⁹³, but the potential risks and
770 limitations of both the torrefaction process and torrefied biomass itself should also be explored.

771 The substitution of coal and other fossil fuels with torrefied biomass presents a
772 compelling alternative for mitigating the environmental impacts associated with energy
773 production. Co-firing torrefied biomass alongside coal, for example, has been shown to
774 significantly decrease greenhouse gas emissions from power plants.⁹⁴ Torrefied biomass
775 typically exhibits higher heating values and calorific potentials than those of raw biomass,⁹⁵
776 making it a more efficient energy source. This enhancement in energy density not only
777 improves combustion efficiency but also expands the range of applications for biomass-derived
778 fuels in various industrial processes, which can contribute as a solution in transitioning towards
779 cleaner energy systems. In addition, biomass torrefaction offers an alternative method for
780 repurposing agricultural residues, such as the energy densification of sugarcane bagasse, as
781 demonstrated in the study by Conag *et al.*⁸², and the improvements in biomass powder
782 flowability for different biomasses (soybean straw, corn straw, rice straw, and rice husk), as
783 explored by Xu *et al.*⁸⁴.

784 Torrefied biomass offers several advantages over raw biomass, particularly in terms of
785 transportation, storage, and densification.⁹⁶ Due to its reduced moisture content and improved
786 bulk and flow properties resulting from the torrefaction process, such as increased energy
787 density and enhanced grindability, torrefied biomass exhibits higher stability during long-
788 distance transportation. This stability minimizes the risk of particle degradation and ensures
789 consistent quality upon arrival at its destination, making it more economically viable and
790 practical for large-scale energy production facilities. The densification of torrefied biomass into

791 pellets or briquettes becomes more efficient and cost-effective, further streamlining handling
792 and storage processes. These characteristics improve logistical efficiency and facilitate the
793 integration of biomass-based fuels into existing energy infrastructure, promoting sustainable
794 energy solutions with reduced environmental impact. In addition, the evaluation of torrefaction
795 pre-treatment on pellet durability with and without binders is also an important topic for
796 efficient feeding of these materials to reactors that needs further investigation, particularly to
797 feed more materials while avoiding issues of pellet breakage or segregation.⁹⁷

798 The application of torrefied biomass can significantly enhance soil properties, offering
799 benefits such as increased water-holding capacity and improved nutrient availability.^{93,98} While
800 torrefaction increases the hydrophobicity of biomass, water-holding capacity is enhanced due
801 to the formation of a porous structure of micro- and mesopores (interporosity) and within the
802 torrefied biomass particles (intraporosity)^{99,100}. However, as explored by Edeh and Mašek¹⁰¹
803 biochar hydrophobicity increases with decreasing particle size, leading to a reduction in water
804 retention capacity; they recommend using hydrophilic biochars with high intraporosity to
805 enhance water retention in coarse soils. In this sense, torrefied biomass can reduce hydraulic
806 conductivity, decrease water losses due to evaporation, and make water more available for plant
807 uptake¹⁰². This enhanced water retention capability not only reduces irrigation needs but also
808 promotes better moisture management¹⁰⁰, particularly in arid or drought-prone regions.

809 Torrefied biomass releases nutrients slowly over time as it decomposes, providing a
810 steady supply of essential elements like nitrogen, phosphorus, and potassium to support plant
811 growth.⁹² These nutrients act as natural fertilizers, enhancing soil fertility and productivity
812 without the immediate risk of leaching or runoff, thereby contributing to sustainable agricultural
813 practices. In contaminated sites, torrefied biomass can be a potential cost-effective adsorbent
814 for the retention and sorption of contaminants.^{103,104} Its ability to effectively bind pollutants
815 makes it promising for the remediation and reuse of organic wastes and residues. Despite these

816 advantages, the scalability of these applications requires thorough investigation, as well as an
817 understanding of the long-term effects of torrefied biomass application in soils. Potential
818 desorption of contaminants over time can hinder the environmental sustainability and
819 effectiveness of remediation strategies. Further research is needed to explore these aspects,
820 ensuring the proper application of torrefied biomass in environmental remediation.

821 The utilization of torrefied biomass, while offering several environmental benefits, also
822 presents potential risks that require critical assessment. The selection of the feedstock for
823 producing the torrefied biomass should consider location factors and logistics, considering
824 biomass residues largely produced and available near the processing units. The energy input
825 required for the torrefaction process should be considered for assessing the carbon savings
826 achieved during combustion. Improper disposal or land application of ash could lead to soil
827 contamination and nutrient imbalances, impacting local ecosystems and agricultural
828 productivity. Furthermore, the long-term impacts of torrefied biomass application in soil
829 remediation and agriculture still need further research, as the torrefied biomass itself can be
830 toxic for several organisms.¹⁰⁵

831

832 **4 Final considerations**

833 From this systematic review on biomass torrefaction, it was observed that most papers
834 focused on determining the chemical and thermal properties of raw and torrefied biomass (62%)
835 to maximize its energy density and solids yield. In contrast, only some studies (8%) presented
836 a complete characterization of physical and bulk properties, and less than 2% investigated
837 flowability or fluidization characteristics for the raw and torrefied biomass, which is critical for
838 the reliable operation of processes at the pilot to industrial scales. This highlights the need for
839 future research on the flowability of raw and torrefied biomass, with the goal of obtaining more

840 sound and broad conclusions for the flow behavior of such heterogeneous materials,
841 culminating in the development of standardized protocols to improve biomass handling and
842 processing at an industrial scale. Some considerations for future studies are summarized below:

843 - No paper has performed a complete characterization of raw and torrefied biomass
844 encompassing physicochemical, thermal, bulk, and flow properties, a surprising feature
845 that should be covered in future studies.

846 - Most experimental works were based on the torrefaction of limited amounts of biomass,
847 on the order of a few grams, which can be limiting for extrapolation or interpretation of
848 flowability results to industrial scale process conditions, particularly because at this
849 latter scale other bulk solids challenges that can prevent powder flow may arise, such
850 as more pronounced powder segregation, effect of powder bed resistances, effect of
851 pipeline defects and wear, etc. Therefore, future studies should assess bulk and flow
852 properties from large-scale torrefaction units and compare them with small-scale ones.

853 - Concerning the flowability characterization, current works focused more on simple
854 classical methods based on powder compressibility or static angle of repose, while very
855 few employed shear cells or rotating drums. Moreover, none among the (few) studies
856 on biomass torrefaction provided a full picture of flowability characterization using all
857 the available techniques. Thus, future studies could cross-check different flowability
858 techniques for a range of raw and torrefied biomass and verify the reproducibility of
859 results by using different equipment brands of a similar measurement technique.

860 - Currently, the studies focused more on the characterization of wood and agro-industrial
861 biomass wastes, however, in the future more heterogeneous and complex wastes, such
862 as municipal wastes or refuse-derived fuels, should be investigated concerning
863 chemical, physical, bulk, and flow properties, aiming at proposing successful solutions
864 for thermochemical valorization of such materials.

865 **Disclosure statement**

866 The authors declare no competing financial interest.

867

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871

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898 new processes for bioenergy production, which pave the way for a more sustainable future.

899

900 **Authors contributions**

901 **Lucas Massaro Sousa:** Conceptualization, Methodology, Formal analysis, Investigation,
902 Writing – Original draft, Reviewing and Editing; Supervision; Project Administration; **Allan**
903 **Pretti Ogura:** Methodology, Formal analysis, Investigation, Writing – Original draft,
904 Reviewing and Editing; **Chayene Gonçalves Anchieta:** Methodology, Formal analysis,
905 Investigation, Writing – Original draft, Reviewing and Editing; **Mathieu Morin:** Writing –
906 Reviewing and Editing; **Nicholas Slongo Canabarro:** Conceptualization, Methodology,
907 Formal analysis, Investigation, Writing – Original draft, Reviewing and Editing.

908

909 **Abbreviations**

910 α : static angle of repose; β : angle of repose; AFM: atomic force measurement; ASTM:
911 American Society for Testing and Materials; AWL: Anhydrous Weight Loss; CCI: Carr's
912 compressibility index; CHNS/O: carbon, hydrogen, nitrogen, sulfur, and oxygen ratio; CT:
913 computed tomography; FFC: flow function coefficient; FTIR: Fourier Transform Infrared
914 Spectroscopy analysis; NMR: nuclear magnetic resonance; H/C: hydrogen/carbon ratio; HR:
915 Hausner ratio; ICP-MS: inductively coupled plasma-mass spectrometer O/C: oxygen/carbon
916 ratio; PRISMA: Preferred Reporting Items for Systematic Reviews and Meta-Analyses; RSM:
917 Response Surface Methodology; SAXS: X-ray-based synchrotron-based scattering; SEM:
918 scanning electron microscopy; TGA: Thermogravimetric analysis; XPS: X-ray Photoelectron
919 Spectroscopy; XRD: X-ray diffraction analysis; XRF: X-ray fluorescence.

920

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